

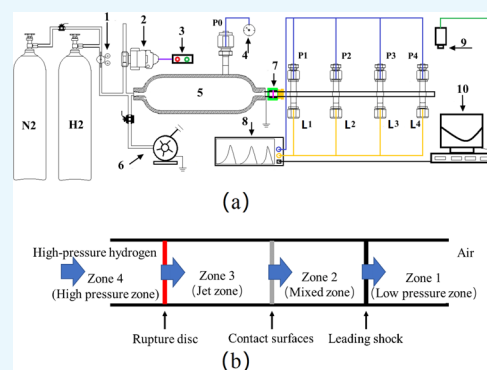
Experimental Investigation of the Self-Ignition and Jet Flame of Hydrogen Jets Released under Different Conditions

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ABSTRACT: Hydrogen is a promising clean energy source and an important chemical raw material. To use hydrogen energy more safely, a high-pressure hydrogen-release platform for hydrogen self-ignition and for generating hydrogen jet flames under different experimental conditions was investigated in this study. The associated experimental analysis was based on the theory of high-pressure hydrogen tube diffusion. We found that the higher the initial release pressure, the greater the intensity of the leading shock. When the initial release pressure was high and the leading shock intensity was strong, hydrogen was more likely to ignite spontaneously inside the tube. The higher the initial release pressure, the faster the average propagation speed of the shock in the same pipe length. The time during which a stable leading shock was formed inside the tube may be related to the initial release pressure. It was found that flame combustion intensified after the passage of air through a Mach disk, and a stable flame was formed more easily at the jet boundary layer away from the orifice axis. The maximum speed of the flame tip and the flame decay speed were very high. Moreover, the flame length and the diameter of the ball flame first increased and then decreased.



1. INTRODUCTION

Hydrogen has a high calorific value of combustion, and it is one of the most important clean energy sources. Many developed countries believe that it is necessary to develop hydrogen fuel cells and the hydrogen industry.¹ However, the minimum ignition energy of hydrogen (0.017 mJ) is extremely low, and its spark ignition energy of explosion is approximately 0.006 J.² Moreover, the limit of hydrogen combustion is very wide, ranging from 4 to 75%. Hydrogen is stored primarily under high pressure. High-pressure hydrogen can easily cause a fire and explode in the event of a leak.³ Therefore, the safety of hydrogen is a major challenge from the viewpoint of promoting its widespread use.

In a leak, high-pressure hydrogen is released into air through a downstream pipeline, and a shockwave is formed at the front of the hydrogen jet. The high temperature generated by the shockwave and the multidimensional shock structure can heat the air in the area through which the shockwave passes. Subsequently, a mixed layer is formed by the hydrogen jet and the aforementioned high-temperature air. When the hydrogen concentration of the layer is in the ignition range and the high temperature of air reaches the hydrogen self-ignition temperature, hydrogen ignites spontaneously according to the diffusion ignition theory of high-pressure hydrogen self-ignition. Duan et al.⁴ found that the higher the pressure of hydrogen release, the more intense the resulting shockwave and the easier the spontaneous ignition of hydrogen. Grune et

al.⁵ studied the spontaneous ignition of leaking high-pressure hydrogen by using transparent circular pipes and schlieren technology. They found that a multidimensional reflection shockwave structure developed between the front of the hydrogen jet and the front shockwave. Duan et al.,⁴ Lee et al.,⁶ and Mogi et al.⁷ have expressed the same viewpoint in their papers regarding the relationship between the pipe length and hydrogen self-ignition; that is, as the pipe length increases, the possibility of hydrogen self-ignition increases, and the minimum release pressure of hydrogen self-ignition decreases. Frederick et al.⁸ determined that the necessary conditions for hydrogen self-ignition are the presence of a combustible mixture of air and hydrogen inside a pipe and the heating of the mixture by the shockwave to reach the hydrogen self-ignition point. This implies that spontaneous ignition of hydrogen is difficult in the absence of a pipeline. Kitabayashi et al.⁹ studied the spontaneous ignition of high-pressure hydrogen venting from pipes of different lengths. They found that the initial release pressure at which high-pressure hydrogen self-ignites can increase when the pipe length exceeds 1.2 m.

Kaneko et al.¹⁰ used a rupture disk to release high-pressure hydrogen into a pipeline and a transparent window at the end of the pipeline to observe the blasting of the rupture disk. They

Received: April 28, 2019

Accepted: May 29, 2019

Published: July 10, 2019

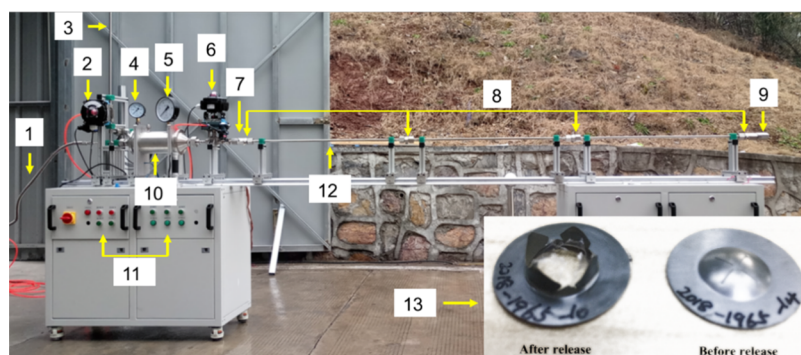


Figure 1. High-pressure hydrogen-release platform: (1) inflatable pipeline; (2) control vent valve; (3) vent tube; (4) vacuum gauge; (5) pressure gauge; (6) emergency brake valve; (7) position of installed rupture disk; (8) sensor locations; (9) nozzle; (10) storage tank; (11) control cabinet; (12) release tube; and (13) rupture disk.

proposed that the blasting process was the main cause of hydrogen self-ignition. In another study, Kaneko et al.¹¹ used a transparent downstream pipe to observe hydrogen self-ignition inside the pipe. In this experiment, they found that hydrogen self-ignition depends heavily on the intensity of the shockwave generated as a result of breakage of the diaphragm. Erez et al.¹² considered that the contact surface may be created because of the sudden release of high-pressure hydrogen and air after breakage of the rupture disk, and turbulence is formed on the contact surface to promote the mixing of hydrogen and air. This provides favorable conditions for the spontaneous ignition of hydrogen.

Studer et al.¹³ conducted large-scale experiments to measure experimental parameters, such as the length of the jet flame, heat radiation flux, and overpressure after hydrogen evolution. Mogi et al.¹⁴ found that the location of hydrogen self-ignition inside the pipe was closer to the diaphragm at higher blasting pressures. Moreover, they observed changes in the flame shape at the nozzle by using a high-speed camera. Duan et al.¹⁵ employed a protective box at the nozzle to observe the flame shape and applied the schlieren technique to observe typical pressure wave structures, such as the Mach disk structure at the nozzle. Mogi et al.¹⁴ and Duan et al.¹⁵ have studied the evolution of the flame morphology at the nozzle, but these researchers have not conducted in-depth research on the flame morphology, such as the length, width, and speed of the jet flame, associated with the spray of a high-pressure hydrogen flame into large spaces.

The experiment of Kitabayashi et al.⁹ lacked direct data on the development of shock and flame inside the tube. To grasp the attenuation process of the shock wave and the development process of the flame inside the tube, this paper discusses experimental research in this area. In this experiment, photoelectric sensors were installed at different locations of the tube to monitor the position of hydrogen spontaneous combustion inside the tube. High-pressure hydrogen is released by pressurization to make the rupture disk naturally rupture, which is more similar to the sudden release of high-pressure hydrogen in reality. The main research objectives of the present work are as follows:

Experimental analysis based on measurement data obtained using pressure sensors, photoelectric sensors, and diffusion theory of spontaneous hydrogen ignition inside a tube;

Experimental study of morphological changes in the external jet flame formed by hydrogen self-ignition inside the tube;

Experimental investigation of hydrogen self-ignition inside the tube and the resulting jet flame outside the tube under different pipe lengths and different initial release pressures.

2. EXPERIMENTAL SETUP

The present experimental study was based on an experimental high-pressure hydrogen release platform, and the schematic of the experimental device is shown in Figure 1. This experimental platform was composed mainly of a cylinder group, a high-pressure storage tank, a holder with a rupture disk, high-pressure discharge tubes, a vacuum pump system, and pressure and photoelectric measurement systems. The high-pressure storage tank included one hydrogen cylinder and one nitrogen cylinder. Hydrogen (99.999%) was the experimental gas. Nitrogen (99.999%) was the test gas in the early experimental stage, and it was used mainly to conduct an airtightness test before the experiment and to purge tanks and tubes after the experiment. The hydrogen tank, emptying tube, and other steel devices were made of 316L stainless steel. The pressure capacity of all devices comprising the experimental setup was 30 MPa.

The pressure sensors and photoelectric sensors were installed symmetrically. Each long tube comprised three short tubes of equal length that were connected using the inbuilt thread structure (see Figure 2), and the inner diameter

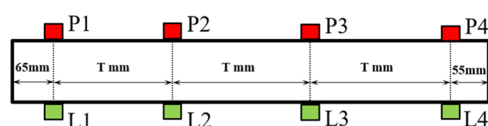


Figure 2. Schematic of tube length and sensor arrangement.

of the tubes was 10 mm. The sensors were installed at four locations on the long tube; P1–P4 were pressure sensors (PCB-113B22), and L1–L4 were photoelectric sensors (Thorlabs FDS-010). An oscilloscope (Agilent-N9003A) and a Keysight high-speed acquisition card (U2531) were used to acquire data from the pressure sensor and the photoelectric sensor, respectively. The natural blasting method was applied to the rupture disk to release hydrogen. The rupture disk was embedded with a “cross” that served as a weakening groove; this structure of the rupture disk produced the least debris during blasting. The evolution of the spontaneous ignition of high-pressure hydrogen to form a jet flame was studied under different initial release pressures and pipe lengths. The lengths

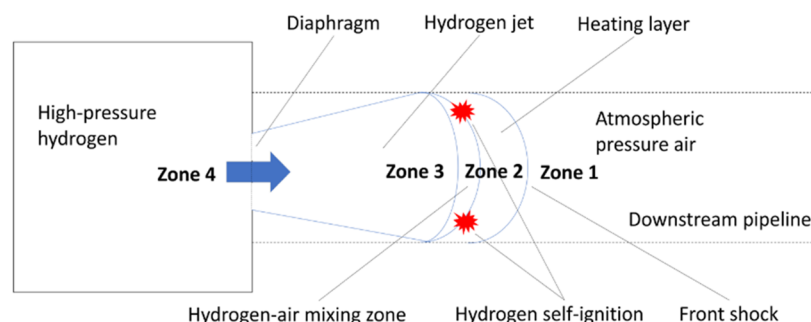


Figure 3. Theoretical diagram of high-pressure hydrogen diffusion inside a tube.

of the experimental tubes were 300, 700, 1200, 1700, and 2200 mm (namely, 3T + 65 mm + 55 mm in Figure 2). The pressure gauge was an industrial pressure gauge, and the accuracy was 0.01 MPa. The degree of vacuum during the experiment is about -1.0 atm. In the experiment, the inlet hydrogen flow rate to the main body container is about $0.6 \text{ m}^3/\text{min}$.

3. RESULTS AND DISCUSSION

3.1. Experimental Analysis Based on the Hydrogen Self-Ignition Diffusion Theory. Figure 3 shows a schematic of the theory of high-pressure hydrogen diffusion inside a tube. The high-pressure chamber inside the tube is filled with high-pressure hydrogen to form the high-pressure zone, that is, zone 4. The low-pressure chamber communicates with the atmospheric environment, and this low-pressure zone is formed by air at atmospheric pressure, that is, zone 1. When the rupture disk blasts suddenly, the sudden release of high-pressure hydrogen from the high-pressure zone forms the leading shock. Because of the turbulent action of the high-speed airflow and the expansion and divergence of the hydrogen jet, the high-pressure hydrogen behind the leading shock mixes with the air in the low-pressure zone downstream of the pipe. A contact surface with the mixed zone is formed in front of the hydrogen jet. The hydrogen–air mixed zone between the leading shock and the contact surface is zone 2. When the initial release pressure is high, the warming effect due to mutual interference of the shockwave compression and the friction between the high-speed airflow and the pipe can increase the temperature in the hydrogen–air mixed zone. When the temperature in the pipe increases and reaches the hydrogen self-ignition temperature and the hydrogen concentration is in the ignition range, hydrogen is ignited after the ignition delay period. The area from the rupture disk to the contact surface is the hydrogen jet zone (i.e., zone 3), and the compressed hydrogen jet is in a state of rapid outward expansion.

Figure 4 shows a graph of the relationship between the initial release pressure and the intensity of the leading shock. The intensity was derived from the stable leading shock detected by the pressure sensors. Xu et al.¹⁶ found that a stable leading shock is formed when the propagation distance (distance from the rupture disk) is $5D$ (D is the diameter of the tube). In the current experiment, the pipe diameter was 10 mm, and the pressure sensor P1 was 65 mm away from the rupture disk. However, we found that the pressure sensor P1 could be used to monitor the stable leading shock in most cases. In some experiments, the stable leading shock must be measured at pressure sensor P2 (detailed description in Figures 5 and 6). By comparing Figures 5b and 6b, the formation of the stable

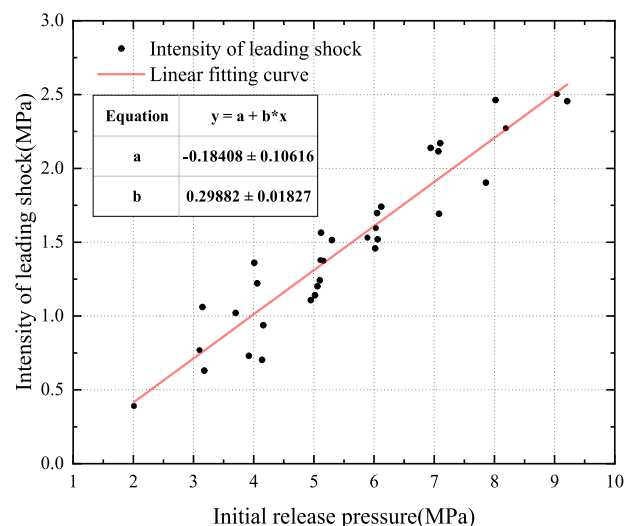


Figure 4. Relationship between the initial release pressure and the intensity of the leading shock.

leading shock was related to the initial release pressure. Figure 4 shows that the greater the initial release pressure, the greater the intensity of the leading shock, and the intensity is distributed above and below the fitting curve.

Figure 5 shows the different experimental conditions tested under a tube length of 300 mm. Figure 6 shows the various experimental conditions tested under a tube length of 700 mm. In Figure 5, for the tube length of 300 mm and an initial release pressure of 4.14 MPa, the stable leading shock could be observed at sensor P1. The initial release pressure was 7.08 MPa, and the stable leading shock could be observed at sensor P2. The leading shock was a strong compression wave, and a series of weak compression waves were superimposed to form the leading shock. Therefore, the formation of the leading shock was a gradual process. During the initial release of high-pressure hydrogen, new compression waves were generated continuously because of the multistep rupture behavior of the rupture disk. When the rupture disk was broken completely, no new compression waves were generated. Thereafter, the intensity of the leading shock remained stable during its propagation. In Figure 6, the maximum leading shock was detected at P2. However, the intensity of the leading shock was unstable. The mutual interference of the shockwave in the propagation process increased the intensity of the leading shock, and energy dissipation because of fluid viscosity and wall friction reduced the intensity of the leading shock (comparing P1 with P4 under one condition). Therefore, the leading shock was relatively stable under the combined action

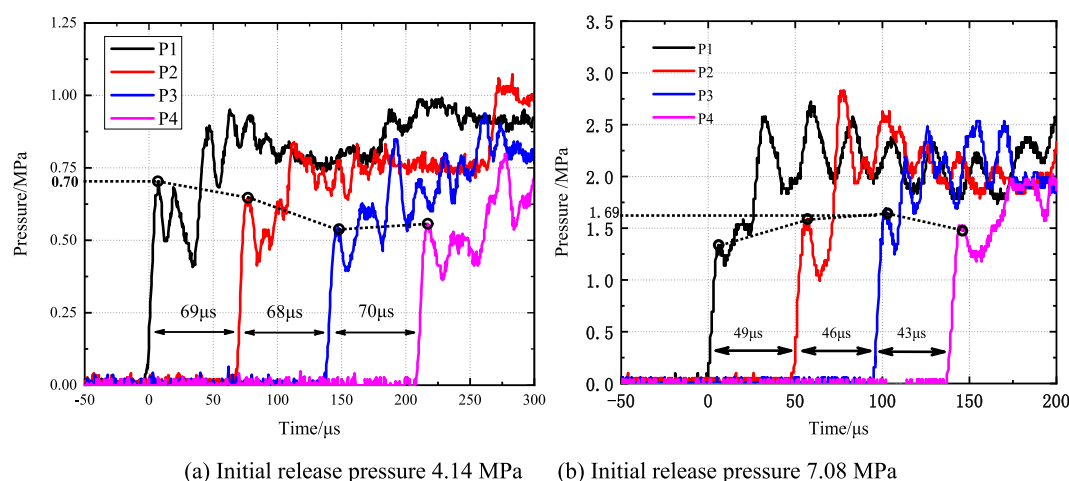


Figure 5. Tube length 300 mm (initial release pressure 4.14 MPa and average speeds of the shock wave 870, 882, and 857 m/s; initial release pressure 7.08 MPa and average speeds of the shock wave 1224, 1304, and 1395 m/s).

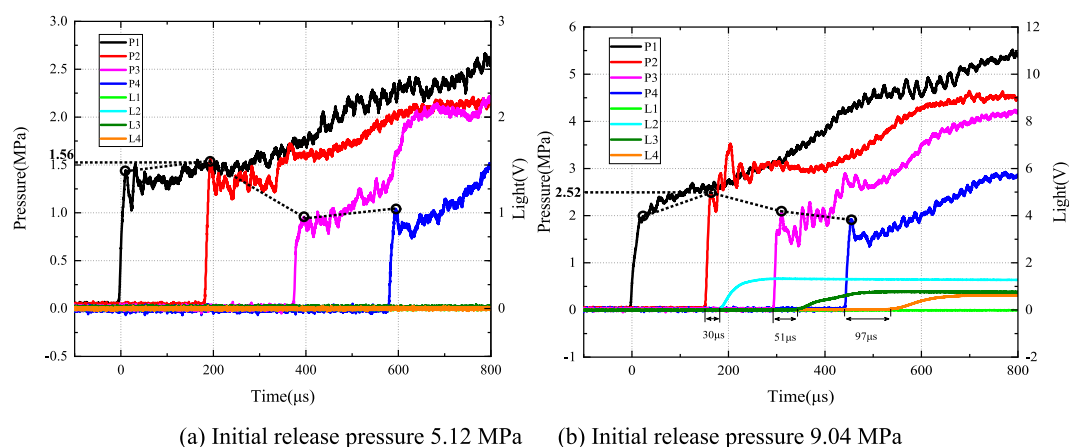


Figure 6. Tube length 700 mm (initial release pressure 5.12 MPa and average speeds of the shock wave 1041, 1007, and 939 m/s; initial release pressure 9.04 MPa and average speeds of the shock wave 1254, 1362, and 1298 m/s).

of the reinforcing effect and the weakening effect. Moreover, Figure 5 shows that the average speed of the shockwave under the initial release pressure of 7.08 MPa was significantly higher than the average speed of the shockwave under the initial release pressure of 4.14 MPa.

In Figure 6b, L2 (photoelectric sensor) first received the photoelectric signal, which shows that the ignition position was between L1 and L2 inside the tube. By comparing the photoelectric signal with the pressure signal, it was found that the leading shock should be in front of the ignition position. Moreover, the interval between the leading shock signal and the photoelectric signal was lengthened gradually, indicating that the mixed zone (zone 2 in Figure 1) enlarged gradually. Kaneko et al.¹¹ found that the self-ignition of hydrogen inside the transparent tube produced a cylindrical flame, and the flame gradually became longer when it propagated downstream. After the rupture disk was broken, the front of the hydrogen jet could form the hydrogen–air mixed zone, and a high-temperature environment could ignite the hydrogen–air mixture. Thereafter, the high-speed hydrogen jet drove the ignited hydrogen and mixed zone toward the tube nozzle. Moreover, the ignited hydrogen–air mixture gradually expanded in the mixed zone.

According to Table 1, when the initial release pressure was low under the same tube length, hydrogen did not ignite

Table 1. Experimental Investigation of Different Initial Release Pressures for the Same Tube Length

tube length 1200 mm						
initial release pressure (MPa)	3.18	3.92	5.02	6.12	7.10	
√self-ignition	×	×	√	√	√	
×non-ignition						
tube length 2200 mm						
initial release pressure (MPa)	2.01	4.16	5.11	6.05	7.86	9.12
√self-ignition	×	×	×	√	√	√
×non-ignition						

spontaneously. Hydrogen self-ignited only after the initial release pressure exceeded a certain threshold. Expansion cooling of the hydrogen jet led to the formation of the hydrogen–air mixed zone (zone 2 in Figure 3) with a temperature that was below the autoignition temperature. This is the main reason that the hydrogen was not ignited easily when the initial release pressure was low. As the initial release pressure increased, the intensity of the leading shock increased and the flow in the hydrogen–air mixed zone (zone 2 in Figure 3) and the hydrogen jet zone (zone 3 in Figure 3) enhanced. Moreover, hydrogen and air mixed more intensely at the contact surface, and shock compression and friction heat

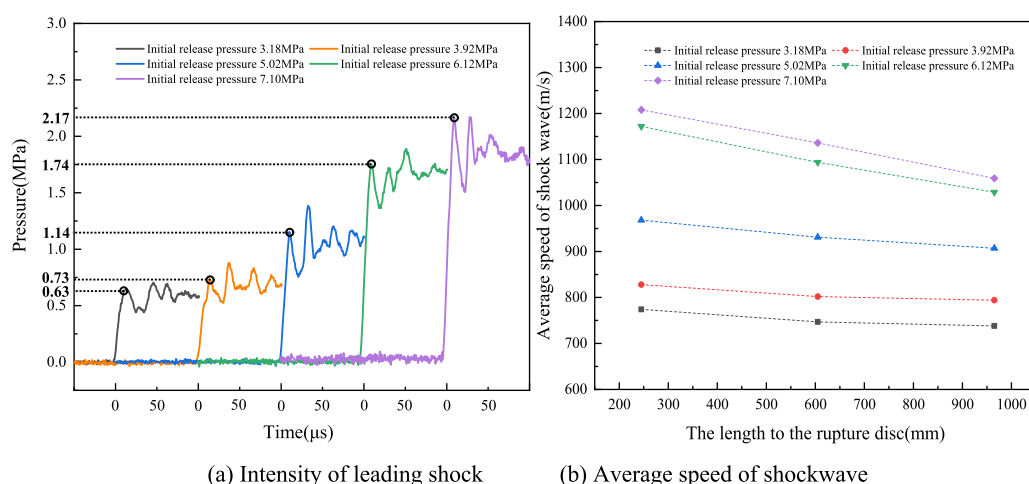


Figure 7. Tube length 1200 mm.

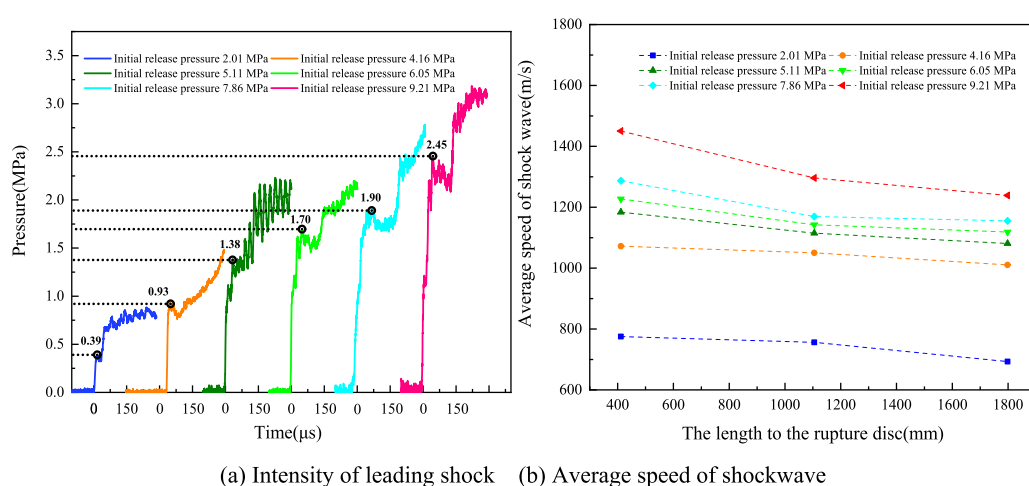


Figure 8. Tube length 2200 mm.

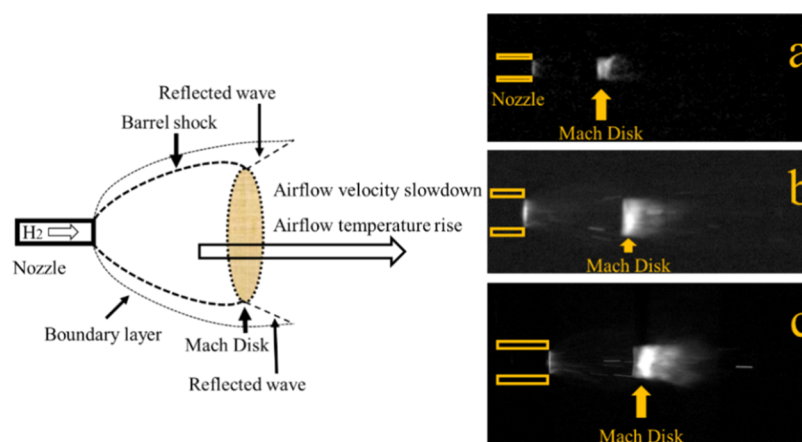


Figure 9. Flame at the nozzle: (a) tube length 700 mm and release pressure 7.08 MPa; (b) tube length 1200 mm and release pressure 6.12 MPa; and (c) tube length 2200 mm and release pressure 7.86 MPa.

generation enhanced, which would eventually promote the self-ignition of hydrogen.

Figures 7 and 8 show the relationship of the initial release pressure with the intensity of the leading shock and the average speed of the shockwave under different experimental conditions. Figures 7 and 8 show that the greater the initial release pressure for the same tube length, the greater the

intensity of the leading shock and the higher the average shock speed. Figures 7b and 8b show that the average speed of the shockwave decreases gradually under the same initial release pressure. This experimental phenomenon was more obvious in the longer tube (compared with Figures 5 and 6) because the shockwave underwent energy dissipation because of fluid viscosity and wall friction during propagation.¹⁷

3.2. Evolution of a Self-Ignition Flame Projecting out of the Tube. Figure 9 shows the flame from the nozzle under different experimental conditions. When the shockwave and the hydrogen jet left the release tube and entered the atmosphere, a complex flow field structure was formed outside the tube. After the leading shockwave passed, the air density increased and the pressure in this area significantly increased. As the high-pressure hydrogen jet entered the atmosphere, it started expanding rapidly. The underexpanded jet developed outward in a supersonic manner at the nozzle. Because the compressed gas had a higher speed at the beginning, which is a momentum-controlled flow, the hydrogen jet propagated substantially along the axial centerline of the tube outlet. Moreover, many expansion waves were generated in the underexpanded jet. The expanding waves propagated outside the jet, and when they encountered the jet boundary layer, they were reflected as compressed waves. The compression waves formed by the reflection propagated toward the inside of the jet and eventually accumulated into a barrel shock in the jet. As the flow continued to develop, shockwaves gradually transformed into a Mach disk. When the barrel shock interacted with the Mach disk, a reflected wave was formed. When airflow passed through the Mach disk, the airflow speed decreased. Therefore, according to the energy conservation law, the temperature of airflow must increase. In Figure 9, when the airflow passed through the Mach disk, the bundle area behind the Mach disk was brighter, indicating that the hydrogen burned more intensely. Thereafter, the flame gradually developed into a slug flame, and, finally, the jet flame was formed. This result is consistent with the experimental results obtained by Kitabayashi et al.⁹ and Kessler et al.¹⁸ A special flame development process was found in these experiments, as detailed in Figure 11.

Figure 10 shows the average speed of the flame tip versus time. In this figure, the average speed of the flame tip

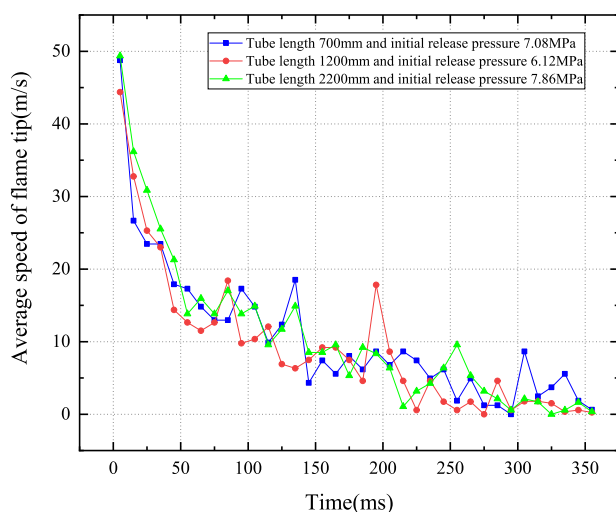


Figure 10. Average speed of the flame tip under different experimental conditions.

decreased gradually with time. In the first 50 ms, the average speed of the flame tip decreased at a faster rate; thereafter, the average speed decreased slowly. In the initial stages of the high-pressure hydrogen jet flame, the initial propagation speed of the flame tip was high because of the high release pressure. From the figure, it can be seen that the initial propagation

speed measured at the flame tip is 45–50 m/s. Then, as the flame outside the tube develops, the speed gradually decreases to 0 m/s because of air resistance.

In the experiment conducted in this study, we used a high-speed camera to record the development of the self-ignition flame projection out of the tube. The frame rate of the high-speed camera was 50 000 fps. Figure 11 shows the flame transition process in the nozzle for the tube length of 2200 mm and an initial release pressure of 9.21 MPa. The time at which the high-speed camera initially captured the flame was set to t_0 . When $t = 0 \mu\text{s}$ in the figure, the self-ignition flame inside the tube was ejected from the nozzle into the external space. Because the high-pressure hydrogen jet started to expand, the flame diameter was greater than the nozzle diameter, and the flame exhibited a fragmented shape. When the flame propagated downstream, the flame surface was broken further. When the hydrogen jet was diffused, the velocity in the central region of the jet was higher and the velocity away from the axis was lower because of the action of the boundary layer. Therefore, at $t = 40 \mu\text{s}$, the flame in the axial region was farther away from the nozzle than the flame at the boundary layer. Because the airflow velocity along the axis was higher, it was difficult for the flame to develop stably. Therefore, at $t = 60 \mu\text{s}$, the flame fragments were blown out at the axis. The airflow velocity at the boundary layer decreased, and the flame stabilized gradually. Because the boundary layer was far from the axis, the interface between the air and the hydrogen jet formed a premixed gas, which provided a sufficient amount of combustible hydrogen–oxygen mixture for the stable flame. As a result, the flame developed rapidly (see flame image at $t = 260 \mu\text{s}$). Subsequently, the flame spread further in the longitudinal direction and at the nozzle, eventually forming the flame at $t = 460 \mu\text{s}$.

Figure 12 shows the development of the out-of-tube flame for the tube length of 2200 mm and an initial release pressure of 9.21 MPa. An out-of-tube flame development map was constructed using the flame image data collected using the digital camera, and the time at which the columnar flame appeared at the nozzle was set to $t = 0 \text{ ms}$. At the beginning of flame formation, the flame inside the tube formed a lumpy flame at the nozzle. Thereafter, the flame developed rapidly. As the high-pressure hydrogen was injected continuously, a bright burning zone was formed. The bright burning zone developed gradually into a ball flame because of jet expansion and divergence, and it moved downstream continuously. At the beginning of the formation of the high-pressure hydrogen gas-spraying flame, the initial release pressure was relatively high, and the jet flame was a momentum-controlled horizontal jet flame.¹³ As the pressure inside the tank continued to decrease, the flame jet weakened. The ball flame kept moving away from the nozzle, and its connection with the jet zone weakened gradually. Then, the ball flame weakened gradually, and the residual flame continued to move downstream for a distance, after which it disappeared. In the later stage of flame development, the flame was controlled by momentum–buoyancy. Finally, the hydrogen inside the tank was exhausted, and the jet flame extinguished gradually. Figure 12 shows that the flame length and width increase first and decrease thereafter.

3.3. Experimental Data of the Self-Ignition Flame Projecting out of the Tube. Table 2 presents experimental results pertaining to the self-ignition flame projecting out of tubes of different lengths under different initial release

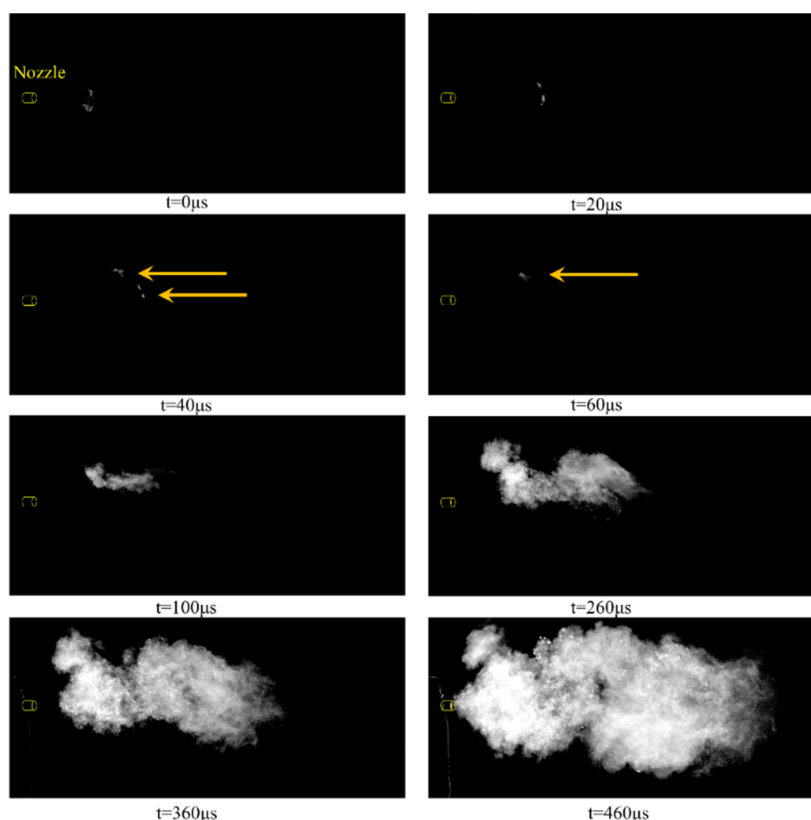


Figure 11. Tube length 2200 mm and initial release pressure 9.21 MPa.

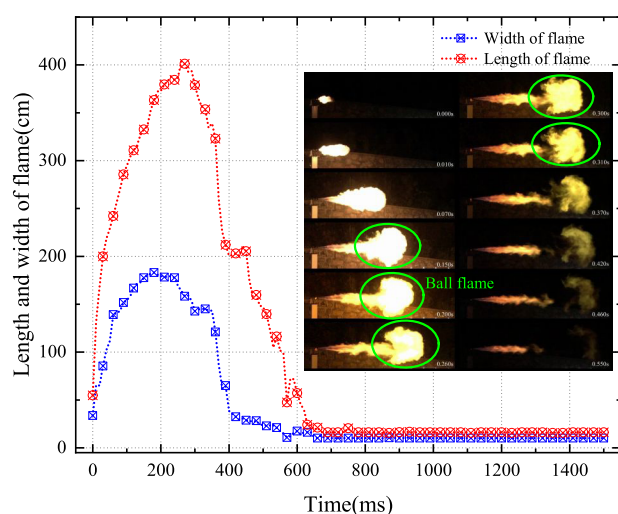


Figure 12. Tube length 2200 mm and initial release pressure 9.21 MPa.

pressures. The maximum shock speed was determined as the average speed of the shockwave inside the pipe between two adjacent pressure sensors. According to the experimental statistics, for the same pipe length, the higher the initial release pressure, the faster the average shockwave propagation speed and the larger the diameter of the ball flame. When the tube length was 1700 mm, the self-ignition position of the hydrogen inside the tube under an initial pressure of 5.79 MPa was closer to the rupture disk when compared with that at an initial release pressure of 3.70 MPa. The diameter of the ball flame did not appear to be regular for different pipe lengths.

4. CONCLUSIONS

In this study, an experiment was conducted to investigate the characteristics of spontaneous ignition and the extra-tube flame in high-pressure hydrogen tubes by changing the tube length and the initial release pressure. The sudden release of high-pressure hydrogen led to the formation of a leading shock, and the intensity of the leading shock was positively correlated to the initial release pressure. The minimum intensity of the leading shock of hydrogen self-ignition inside the tube was approximately 1.0 MPa (conditions: tube length 1700 mm; initial release pressure 3.70 MPa). A comparison of the results obtained under the experimental conditions with tube lengths of 300 and 700 mm showed that the position of the stable leading shock inside the tube was not certain, which might be related to the initial release pressure. In addition, the results obtained under the experimental conditions with tube lengths of 1200 and 2200 mm were compared. The higher the initial release pressure, the faster the average propagation speed of the shockwave. The average shockwave speed decreased gradually because of energy dissipation during downstream propagation. Through high-speed imaging, the flame at the axial position of the nozzle was more likely to be extinguished, and the flame brought to the boundary layer by the vortex was more likely to develop stably. Thereafter, the flame of the boundary layer could ignite the combustible hydrogen–air mixture to form a stable combustion zone. By using a digital camera, it was found that the combustion zone was pushed by the high-pressure and high-speed airflow to form a spherical flame, and the speed of the flame tip reached 76.51 m/s. By reviewing the effective experimental data, it was found that the higher the initial release pressure, the faster the average shock propagation speed, the longer the flame length, and the greater

Table 2. Experimental Investigation of Spontaneous Combustion of Hydrogen under Different Conditions

no.	tube lengths (mm)	initial release pressure (MPa)	maximum shock speed (m/s)	photoelectric signal position	maximum flame tip speed (m/s)	ball flame diameter (m)
1	300	5.83	122	L2 + L3 + L4	34.16	1.43
2	300	7.08	1395	L2 + L3 + L4	48.77	1.75
3	700	5.30	1142	L2 + L3 + L4	40.12	1.51
4	700	6.03	1213	L2 + L3 + L4	40.67	1.75
5	1200	5.02	968	L2 + L3 + L4	30.64	1.49
6	1200	6.12	1172	L2 + L3 + L4	32.76	1.51
7	1700	3.70	1106	L3 + L4	32.08	1.48
8	1700	5.79	1281	L2 + L3 + L4	50.00	1.50
9	2200	7.86	1287	L2 + L3 + L4	49.36	1.83
10	2200	9.21	1450	L2 + L3 + L4	76.51	1.84

the ball flame diameter. However, there was no obvious change in the flame experimental data for different tube lengths.

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Notes

The authors declare no competing financial interest.

ACKNOWLEDGMENTS

This work was supported by the National Key R and D Plan of China (Projects Nos. 2017YFC0804700 and 2016YFC0800100).

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